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Synergistic of ammonium polyphosphate and alumina trihydrate as fire retardants for natural fiber reinforced epoxy composite



P. Khalili ^a, K.Y. Tshai ^a, D. Hui ^b, I. Kong ^{a, *}

- ^a Department of Mechanical, Materials and Manufacturing Engineering, The University of Nottingham, Malaysia Campus, Jalan Broga, 43500, Semenyih, Selangor, Malaysia
- ^b Department of Mechanical Engineering, University of New Orleans, Lake Front, New Orleans, Louisiana, LA 70138, USA

ARTICLE INFO

Article history:
Received 7 September 2016
Received in revised form
11 December 2016
Accepted 28 January 2017
Available online 31 January 2017

Keywords: Hybrid Polymer-matrix composites (PMCs) Mechanical properties Thermal properties Thermosetting resin

ABSTRACT

Various compositions of epoxy/hardener with flame retardant (FR) as additives were used to produce natural fiber composite through the resin infusion technique. The effects of natural fiber (NF), ammonium polyphosphate (APP), alumina trihydrate (ATH) and ATH/APP hybrid on flammability, thermal and mechanical properties of the composites were investigated. Incorporation of NF into neat epoxy matrix reduced the gross heat of combustion with improved thermal degradation properties. Addition of APP enhanced the flame resistant properties of the composite with a much reduced total flame time and zero drip. The 10 wt·% ATH and 5 wt·% APP hybrid showed the most promising flame retardancy with a self-extinguishing property as well as the lowest gross heat and greatest char residue amongst the various formulations investigated. In general, the formulations containing only ATH couldn't provide the effective flame resistivity as compared to that of the APP-filled composites. The ATH-filled formulations reduced the tensile strength, elongation at break and flexural properties while an increase in the flexural strength of APP-loaded formulations was observed. An increase in moduli of the FR-filled composites was measured as compared to that of the NF/epoxy composite, owing to a more brittle characteristic.

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1. Introduction

Over the past decade, the increasing awareness in community health and environment has boosted the utilization of fibres obtained from renewable sources. The usage of natural fiber (NF) composites have been gaining momentum in several applications such as the automotive, furniture and food packaging industries due to their renewability, availability, low density, low cost and appropriate mechanical performances [1–5]. However, the fall-backs such as high flammability behavior of NF, poor compatibility of NF with polymeric matrices of hydrophobic nature, and the high processing temperature of thermoplastic polymers (which adversely affects physio-mechanical performances of NF) [1] broadly limits their wider application, in particularly the venture towards aviation industry which governs by an exhaustive set of stringent regulatory requirements.

E-mail address: ing.kong@nottingham.edu.my (I. Kong).

Unlike thermoplastics, thermoset resin possess low viscosity prior to crosslink and their fabrication route does not require to be operated at high temperature. The merit of thermosetting polymer provides ease of fabrication for accommodating constituent mixture of NF reinforced composites [6], and it was also found that epoxy resin could exhibit stronger surface interaction than that of the unsaturated polyester. In addition, processing of thermosetting composites can be performed with much lower processing pressure than that of the thermoplastic polymers [7].

Intumescent flame retardants (IFRs) have been widely used to reduce the flammability of polymers, with enhanced properties such as low corrosion and toxicity, reduced smoke emission, increased char formation and anti-dripping flame [8]. IFRs consist of a source of blowing agent, a source of char formation, and an acid source which together capable of generating a foamed char barrier that decreases the transport of fuel and heat into the bulk of the constituents [9–11]. In the event of fire, an intumescent ammonium polyphosphate (APP) agitated by the heat source eliminate ammonia (NH₃) and water while forming ultraphosphate which could act as a protective layer against fire propagation. Alumina trihydrate (ATH) on the other hand undergoes endothermic

^{*} Corresponding author. Department of Mechanical, Materials and Manufacturing Engineering, The University of Nottingham, Malaysia Campus, Jalan Broga, 43500, Semenyih, Selangor, Malaysia.

dehydration upon fire, releasing water vapor which function as diluent for the combustible gases with concurrent formation of a thermally stable ceramic alumina (Al_2O_3) that serve as an insulating layer against heat and mass transfer [12,13]. The chemical structures of APP and ATH are shown in Fig. 1. However, increasing the concentration of FRs in a polymeric formulation often associated with reduction in mechanical performances and inevitably increased material cost, thus the smallest effective FR loading, which could be up to 20 wt·%, is generally desirable [14]. From the manufacturers' perspective, it is crucial to derive the optimum amount of IFRs to meet the demand for fire retardant system.

Castronvinci et al. [15] investigated the effects of APP/ATH hybrid on fire retardancy of styrene butadiene rubber (SBR) block

Fig. 1. The chemical structures of (a) APP and (b) ATH.

copolymer noticed clear interactions between APP and ATH as a FR system. The chemical reactions took place over a varying range of temperatures. The chemical reaction first occurred between APP (NH₄PO₃) and hydroxyl groups (-OH) of ATH at a temperature lower than 400 °C (Fig. 2). This resulted in the formation of aluminum phosphate with generation of water and ammonia (NH₃) out of ammonium salt (NH₄). These reactions kept repeating until aluminum metaphosphate ([Al(PO₃)₃]_n) was formed.

At higher temperature, further reactions took place forming aluminum orthophosphate (AlPO₄) from the long chain polyphosphate as well as releasing ammonia and water (Fig. 3).

The combined effect of APP/ATH contributed to an improved fire extinguishing behavior due to a number of factors. Firstly, the generation of aluminum phosphate prevents the evaporation of phosphorous volatility. Secondly, the basicity of ATH that furnish protons exchange and an easier evolution of water and ammonia gas. Thirdly, the crystalline form of Al-P-O alters the glassy ultraphosphate structure by which gases can readily escape the solid surface.

Introduction of NF into neat polymers reduced the heat release and enhanced flame retardant performances relative to those of the pure matrices [16–18], and incorporation of small loading level of FR could further reduce the flammability behavior of biocomposite at the cost of a slight drop in mechanical properties. Few reports

(1)
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Fig. 2. Reactions between ATH and APP.

Fig. 3. Reactions between ATH and APP at elevated temperatures.

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